

*The Photographic Action of the α -Particles emitted from Radio-active Substances.**

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It has been shown by Rutherford† that the photographic action of α -particles stops suddenly when the thickness of an interposed aluminium screen is increased beyond a certain value. It is noticeable that the thickness of the screen required to completely stop photographic action is identical with that required to stop ionisation‡ in gases and scintillations§ of a phosphorescent screen caused by α -particles.

The first object of the present experiments was to investigate the manner in which the photographic action of α -particles varies along their path, and to see if there is any connection between this and the well-known variation of the ionisation produced in gases. This having been accomplished, a relation was found between the photometric density of the film produced on development and the number of α -particles incident upon it. Experiments were then made to see whether a single α -particle produced a detectable photographic effect.

We define the photographic action of the α -particles on a sensitive film as the relative amount of the silver halide altered under the influence of the particles and thus rendered capable of development. A measure of the action is therefore the photometric density $D_{||}$ of the film on complete development, this quantity being proportional to the amount of silver contained in the film.¶

Hurter and Driffeld** have shown that if D_t is the density after development for time t , then

$$D_t = D_{\infty} (1 - a^t),$$

* A brief account of this paper was communicated to the meeting of the British Association at Winnipeg.

† 'Phil. Mag.,' July, 1905, Jan. and April, 1906.

‡ Bragg and Kleeman, 'Phil. Mag.,' Dec., 1904, and Sept., 1905.

§ Rutherford, 'Phil. Mag.,' vol. 12, p. 145.

|| As is usual, D is defined as $\log_{10} I_0/I$, where I_0 is the intensity of the incident and I that of the transmitted light.

¶ Hurter and Driffeld, "Photog. Journ.," 1898; Eder, 'Beiträge zur Photochemie u. Spektral-Analyse,' 1904; Luther, 'Zeit. f. Phys. Chem.,' 1900; Sheppard and Mees, 'Roy. Soc. Proc.,' p. 452, 1905.

** 'Journ. Soc. Chem. Ind.,' May, 1890.

D_{∞} being the limiting density which would be reached by infinitely long development and a a constant. Therefore if exactly similar films are developed under identical conditions, the density D will in each case be proportional to the limiting density D_{∞} , which gives the magnitude of the change in the film.*

After repeated failures to obtain concordant results with ordinary commercial plates and developers, it was found essential to use special plates and to use great precautions in developing. In the following experiments, Wratten's "Instantaneous" and "Ordinary" emulsions thinly coated on patent plate glass were employed, the central portion only of the plate being used. Except where otherwise mentioned, all the plates were developed in a $\frac{1}{16}$ N solution of ferrous sulphate in 2 N potassium oxalate for 20 minutes, which time was found to be sufficient for even development throughout the thin film used.

Each series was done simultaneously, so that the temperatures of development were the same. The temperature, being that of the room, varied only between 15° C. and 17° C. on different occasions. Density measurements were made with a König's spectro-photometer, green light being always used. Thus, for films from the same plate the density was proportional to the amount of silver halide altered.

Now the α -particles emitted by ordinary radio-active substances are heterogeneous, and are consequently not suited for quantitative experiments of this nature. Those emitted by a thin layer of radium C, however, are homogeneous and have an initial velocity of 2.06×10^9 cm. per second.† The number of the particles falling on a known area of the film during an exposure can be calculated from the known decay curve of radium C. provided that the γ -ray activity of the radium C is once determined in terms of a radium standard.

Suppose that a narrow beam of the homogeneous and evenly distributed α -rays from radium C falls on a film of thickness x_0 normally to its surface with their initial velocity. Provided that the film is uniformly made and developed, the density δD which would be produced in a very thin layer of the film between two planes at distances x and $x + \delta x$ respectively from the surface will be a function of N and v , N being the number of α -particles which have passed through unit area of this layer and v the velocity of the particles in the layer.

* It must be remembered that this is only true for a very thin film, of which we can develop the surface and bottom to an equal extent, and *does not hold* for a thick film, which it is impossible to develop evenly, even in the course of two or three hours.

† Rutherford, 'Phil. Mag.', Jan., 1907.

Since the velocity of the particles would be uniform in the layer, we can take x as an independent variable in place of v . Thus $\delta D = f(N, x) \delta x$, and the *density* of the film, which we can measure by means of a spectrophotometer, is its integral value, as

$$D(N, x_0) = \int_0^{x_0} f(N, x) dx.$$

Since in the case of ordinary light the intensity of the light decreases through the film according to an exponential law with the distance traversed, the relation between the exposure and the density produced can be determined without taking much account of the thickness of the film, so long as it is the same in each case. In the present experiments, however, if the thickness of the film is less than the range of the α -particles in the substance, the particles can pass through it and still have photographic action.

If the rays are screened by a layer of matter of uniform thickness before they reach the surface of the film, the photographic action may stop in the film at a certain distance from the surface, this distance depending on the velocity of the incident rays. Consequently we have to take the actual thickness of the film into consideration, and to deal with $\partial D / \partial x$ or $f(N, x)$, which is proportional to the actual photographic action of N α -particles with the velocity corresponding to the distance x , previously traversed by the particles.

For convenience we shall call $f(N, x)$ the "*differential density*" at distance x . $f(N, x)$ has a meaning also in non-sensitive substances like aluminium or air, if we take it to mean the photographic action which would be produced on a sensitive film after the α -particles had traversed a thickness x of these substances.

Variation of the Photographic Action with the Thickness of Aluminium Screens.

The apparatus employed for this purpose is shown in section in fig. 1.

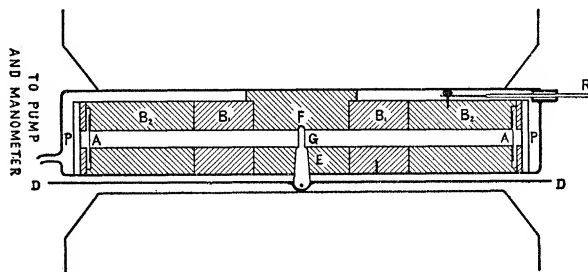


FIG. 1.

B_1 and B_2 are solid concentric brass cylinders through both of which 10 holes of 5 mm. diameter are radially bored. The holes can be covered with any desired number of aluminium sheets of uniform thickness at the outer ends A, over which the photographic plates P, 1.3 cm. sq., are placed, the distance of the plates from the axis of the cylinders being 4.9 cm. These two cylinders fit in a metal box over the circular projection F which forms a part of the box. When the box is sealed, a circular steel rod G, of diameter 2.3 mm., which is coated with the active deposit from the radium emanation, goes through a central hole in F and can be kept along the axis of the cylinders by means of an air-tight conical joint.

In order to prevent the α -rays from the rod acting on the photographic plate before all is ready, the external cylinder B_2 is turned so that the holes in B_1 are closed. One end of G fits into a disc D (outside the box) by which it may be kept in rotation. The whole apparatus is brought into a magnetic field to deflect the β -rays to the walls of the holes. After evacuating the box to a pressure of 2 or 3 mm. of mercury with a Fleuss pump, the holes are opened without changing the internal pressure, by turning B_2 with the rod R, which projects through a hole in the box, B_1 being kept fixed by a plug. The α - and γ -rays are then in a position to act on the photographic plate.

When the necessary exposure has been given, B_2 is again turned to the closed position. It was found that when G was not rotated during the exposure the densities of the photographs taken without any screen were not the same, owing to the non-uniform distribution of the active deposit on G, the maximum difference being sometimes 25 per cent. This was reduced to about 2 per cent. when G was constantly rotated during an exposure.

The holes were then covered with 0, 1, 2, . . . 9 sheets of aluminium foil, each weighing 0.00075 gramme per square centimetre. When the experiments were finished with these numbers they were altered to 0, 1, 8, 9, . . . 16, and a similar series of experiments was performed. In each case the exposure commenced at least half an hour after the removal of G from the emanation vessel, so that the number of α -particles emitted from radium A was negligible compared with that from radium C.

Table I gives the experimental results, these being the average of several experiments.

They are shown graphically in Curves I and II in fig. 2, the abscissæ representing the number of the aluminium sheets and the ordinates the corresponding densities.

It can be seen that D first remains constant as σ , the number of aluminium

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 sheets, increases, but begins to decrease at $\sigma = 8.5$ for the "Instantaneous"
 plates and at $\sigma = 10.7$ for the "Ordinary" plates, in both cases vanishing at
 $\sigma = 14$.

Table I.

Number of aluminium sheets, σ .	Density D, in relative values.	
	"Instantaneous" Curve I.	"Ordinary" Curve II.
0	1.03	0.99
1	1.00	0.98
2	1.03	1.03
3	1.03	1.00
4	0.97	0.97
5	0.98	0.98
6	0.98	1.02
7	0.98	0.98
8	1.01	1.02
9	0.96	1.02
10	0.73	0.99
11	0.54	0.95
12	0.40	0.65
13	0.15	0.26
14	0.00	0.00
15	0.00	0.00

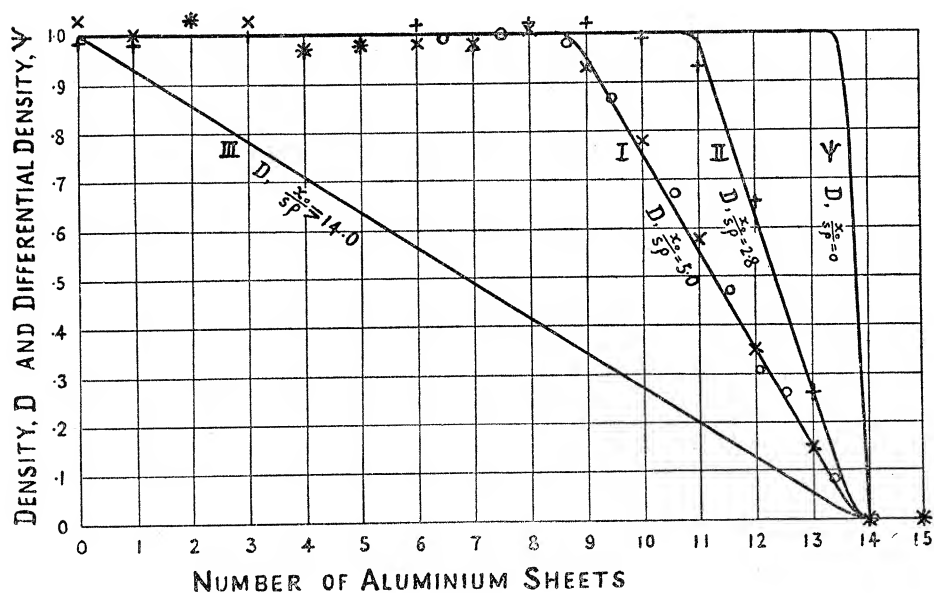


FIG. 2.

The general character of these curves remained the same for any value of N , and therefore the expression for D assumes the form*

$$D(N, x_0) = \phi(N) \int_0^{x_0} \psi(x) dx, \quad (1)$$

where $\phi(N)$ is a function of N only and $\psi(x)$, that of x only, being proportional to the "differential density."

We have, therefore, to deduce the latter from the experimental D curves, which are different for different plates, taking the thickness of the film and the relative retarding powers of the aluminium and the film into consideration.

If the rays are screened by σ sheets of aluminium foil, the velocity of the α -rays incident on the film is decreased to the value corresponding to that at $x = \sigma s\rho$, s being the thickness of each aluminium sheet and ρ the ratio of the retarding power of the aluminium to that of the film.

Therefore, the density of the film corresponding to σ is

$$D(N, x_0, \sigma) = \phi(N) \int_0^{x_0} \psi(x + \sigma s\rho) dx.$$

Differentiating this with respect to σ , we get

$$\frac{\partial D(N, x_0, \sigma)}{\partial \sigma} = s\rho\phi(N) \{\psi(s\rho + x_0) - \psi(s\rho)\}.$$

From the experimental results obtained, D is constant for any value of σ between 0 and 8.5 for the "Instantaneous" plate. Therefore ψ is constant for a range of values of σ between 0 and $8.5 + \frac{x_0}{s\rho}$, where $\frac{x_0}{s\rho}$, or the number of aluminium sheets equivalent to the film, was determined in the following way:—

From other experiments with separable films, made of the same emulsion, it was found that the weight of the film of that thickness at which photographic action stops is 0.0107 gramme per square centimetre. The weight of the film on the "Instantaneous" plate was 0.00382 gramme per square centimetre, so that this film was equivalent to 5.0 sheets of aluminium sheets.

Consequently, ψ is seen to remain constant as σ increases from 0 to 13.5, and then decrease rapidly and vanish at $\sigma = 14.0$. This is shown in fig. 2, curve ψ .

The equivalent number of aluminium sheets of the film on the "Ordinary"

* Since the scattering of the α -rays is small and the density is measured in the central portion of each photograph (the diameter being the same as that of the hole), N can be regarded as independent of x .

plate was similarly found to be 2.8, and the same deduction brings out a similar curve for ψ , as would be expected. Thus D curves depend on the thickness of the film where the "differential density" is the same.

If the film is thick enough to stop the α -particles, the curve D would be like Curve III in fig. 2.

Thickly coated plates were specially prepared to test this point. Contrary to expectation, however, it was found that the D curve could not be obtained, the reason being that we cannot develop the surface and bottom of such thick films to the same extent, even in the course of two or three hours development. The thinner the film, the nearer D will vary as ψ itself.

A very thin collodion film was prepared, but in this case the difficulty of obtaining a perfectly even film made a quantitative measurement of D impossible. The density did not vary much with the number of aluminium sheets from 0 to 12, but fell off abruptly between 12 and 14 sheets, which still supports the above reasoning.

The photographic action of the γ -rays in these observations was found to be negligible, and was practically eliminated as fog.

Variation of the Photographic Action in the Film itself.

Since we cannot develop a thick film uniformly, we have either to cut such a film into very thin layers before development, or to use strata of separable films, and develop each simultaneously.

The first method being impracticable, the second method was adopted.

For this purpose, a thin film was coated on a celluloid sheet from which the film could be detached. Several sheets of these loose films were placed at the end of the uncovered hole in the apparatus described in fig. 1. They were then exposed to the α -rays in a manner similar to that employed in the case of plates and then placed on a wet gelatine plate. When dry, they were developed as usual. The results were rather qualitative in nature, owing to the difficulty of operating and to the unevenness of the loose films—an effect which cannot be avoided.

In most cases, the densities of the first four layers of the films were nearly the same, while the fifth layer had sometimes a very small or zero density, varying with different sheets of the film from which the loose films were taken.

By weighing the film and measuring its area, it was found that the weight of the film which can cut off the photographic action was 0.0107 gramme per square centimetre.

It is, therefore, reasonable to suppose that the variation of the "differential density" is the same as that which we obtained for aluminium, the only difference being the unequal retarding powers of the substances.

Variation with Distance of Photographic Action, using Air at Atmospheric Pressure.

The apparatus employed for this investigation was similar to that shown in fig. 1, except that the distance of plate x from the central axis of the rod G varied as given in Column II, Table II. In this case, therefore, the number of α -particles N_x incident upon unit area of the plate at distance x during exposure will be inversely proportional to x^2 , thus

$$N_x = N_0 x^{-2},$$

where N_0 is the value at unit distance, the measurement of which will be described later.

A series of photographs was taken *in vacuo* and the other in air at atmospheric pressure. The densities D' of the plates observed are given in Columns IV and VII, N_0 being 4.48×10^8 per square centimetre in the former case and 7.84×10^8 per square centimetre in the latter.

Table II.

I	II	III	IV	V	VI	VII	VIII
No.	x , distance in cm.	Vacuo.			Atmospheric pressure.		
		N_x , = N_0/x^2 per sq. cm.	D' , density observed.	D , density reduced to same value of $N = 10^7$ per sq. cm.	N_x , = N_0/x^2 per sq. cm.	D' , density observed.	D , density reduced to same value of $N = 10^7$ per sq. cm.
1	3.23	4.30×10^7	1.46	0.420	7.52×10^7	2.72	0.515
2	3.74	3.20×10^7	1.15	0.407	5.60×10^7	2.24	0.516
3	4.33	2.39×10^7	0.94	0.426	4.18×10^7	1.76	0.507
4	4.72	2.01×10^7	0.79	0.417	3.52×10^7	1.35	0.443
5	5.28	1.61×10^7	0.63	0.406	2.81×10^7	0.88	0.347
6	5.76	1.35×10^7	0.55	0.416	2.36×10^7	0.53	0.243
7	6.03	1.23×10^7	0.50	0.413	2.15×10^7	0.32	0.159
8	6.27	1.14×10^7	0.48	0.425	1.99×10^7	0.25	0.133
9	6.69	1.00×10^7	0.43	0.430	1.75×10^7	0.07	0.042
10	7.04	0.905×10^7	0.38	0.419	1.58×10^7	0.00	0.00

We have, therefore, to deduce corresponding densities D for the same number of α -particles from the observed values.

From another series of experiments, discussed later, it was found that the density of the film "Instantaneous" employed, varied with the number of α -particles as

$$D = 3.7(1 - e^{-1.18 \times 10^8 \times N}).$$

If D is the corresponding value of D' for $N = 10^7$ per sq. cm., then

$$D = D' \frac{1 - e^{-0.118}}{1 - e^{-1.18 \times 10^{-8} \times N_x}}.$$

These are given in Columns V and VIII. Thus the densities of the photographs taken *in vacuo*, when deduced for the same number of α -particles, are the same for each case, within the limits of error of observation. This might have been expected, as there is no absorption. The relative values of the deduced densities given in Column VIII are represented graphically in fig. 2 (the points marked o). For this case, one sheet of aluminium corresponds to 0.5 cm. of air.

It will be seen from that figure that the curve connecting these points* coincides with Curve I, showing that the "differential density" ψ , and therefore the variation of the photographic action along the path of α -particles in air is similar to that in aluminium, the only difference being the unequal retarding powers of the substances.†

The aluminium sheet of sufficient thickness to stop the photographic action has a weight per square centimetre of 0.0105 gramme. In the case of the film the corresponding weight is 0.0107 and for air 0.0091, all the three quantities being of the same order.

Determination of $\phi(N)$ or the Variation of the Density with the Number of α -Particles falling on the Film.

If identical films are exposed to the α -particles emitted by a thin layer of radium C at equal distances from the layer, $\int_0^{x_0} \psi(x) dx$ is constant in each case, hence $\phi(N)$ is proportional to D , as is seen from equation (1).

To investigate the connection between the density of a film and the number of α -particles incident upon it, the apparatus of fig. 3 was used.

A photographic plate P, 40 cm. $\times$ 2 cm., was arranged to slide over the end of a cylindrical hole of 5 mm. diameter made in a metal cylinder, along the axis of which α -rays are expelled from radium C deposited on the cylindrical part of a steel rod G, 2.3 mm. in diameter. This was placed at a distance of 3.76 cm. from the plate. The whole apparatus, being protected from light and placed between the poles of an electro-magnet, only α - and γ -rays could reach the film, while β -rays were deflected to the walls of the hole. By means of a shutter A, the α -rays could be cut off when desired. G was

* The values for distances less than 3.23 cm. were found to be constant. This was done by partially evacuating the box in which the apparatus was placed.

† Similar experiments were made with "Ordinary" plates, but the detail is omitted, the results being the same.

previously exposed to the radium emanation until radio-active equilibrium had been established. If the time at which G is removed from the emanation vessel is known, the number of α -particles emitted by the radium C during the exposure can be calculated from its known decay curve. The total number of α -particles expelled from the radium C was determined by measuring its γ -ray activity with a standardised electroscope, and taking 3.4×10^{10} as the number of the α -particles expelled per second by 1 gramme of pure radium in equilibrium with its products.*

By sliding the photographic plate across the opening a series of photographs (10 to 15) could be taken on one plate, the time t and the interval τ of each exposure being noted.

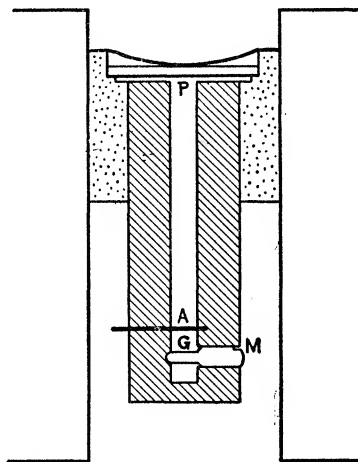


FIG. 3.

We have shown that the photographic action of the α -rays is constant over a range of 6.7 cm. in air. In this case, the film of the plate "Ordinary" was equivalent to 1.4 cm. of air, and that of the plate "Instantaneous" 2.5 cm. These plates being placed at a distance of 3.75 cm. from the radium C, the "differential density" is therefore constant throughout the film in both cases.

The radium C used as the source of α -particles is not constant in amount, but decreases according to the law of successive changes in radio-active substances, and it is not *a priori* certain whether the photographic action can be expressed in terms of the total number of the α -particles, $N = \int_t^{t+\tau} n dt$, which have fallen on the film during the exposure τ , or if it is a function of some function such as $\int_t^{t+\tau} F(n) dt$.

It may be recalled that in the case of light the density is not a function of $I\tau$, but of $I\tau^p$, I being the intensity of the light and p a constant† (about 0.8).

To decide this question, a series of photographs was taken in which the time of exposure τ varied in such a way that the total number N of α -particles

* Rutherford and Geiger, 'Roy. Soc. Proc.' A, vol. 81, p. 156, 1908.

† Schwarzschild, 'Phot. Korr.', 1896, p. 171; Sir W. de W. Abney, 'Phot. Journ.', Oct., 1893; Eder, 'Jahrbuch,' 1899, p. 457.

was the same in each case, while the intensity n was decreasing from its initial value to about 3 per cent. in the course of three hours. The results are given in Table III, the total number N being 1.42×10^7 per square centimetre of the film.

Table III.

Plate, "Ordinary"; pressure, 761 mm.; temperature, 17° C.; $n_0 = 5.69 \times 10^4$.

No.	Exposure.						$\frac{N}{\tau} \times 10^{-4}$.	Photometric reading.	Density, D.
	From—			To—					
	h.	m.	s.	h.	m.	s.			
1	0	10	0	0	15	0	4.72	41.55	0.258
2	0	15	30	0	20	42	4.55	41.6	0.256
3	0	21	0	0	26	42	4.14	41.65	0.255
4	0	27	0	0	33	10	3.83	41.6	0.256
5	0	34	0	0	40	42	3.52	41.7	0.253
6	0	42	0	0	49	35	3.11	41.76	0.251
7	0	50	0	0	59	0	2.62	41.65	0.255
8	1	0	0	1	11	0	2.15	41.6	0.256
9	1	20	0	1	37	58	1.31	41.6	0.256
10	1	40	0	2	13	0	0.72	41.5	0.259

Thus D is constant when N is constant, and is independent of the intensity of the rays so long as N is the same. Consequently the density can be expressed in terms of N for any intensity of the α -rays.

This having been established, experiments were next undertaken to determine the relation between the number of α -particles N and the photometric density of the film produced.

Experiments were performed in a similar manner to those just described, the details of which may be seen in Tables IVA and IVB.

In fig. 4 the densities are plotted against the numbers of α -particles which have fallen on unit area of the film. The points marked (o) represent the results of one series of experiments and those marked ($\times$) and (+) another series. Care was taken to use parts of the same photographic plate when a comparison was required between two series; this was necessary owing to a variation in the thickness of the films on different plates.

The curves actually drawn are theoretical curves to which reference will be made later.

The actual photo-process undergone by the silver halides in the emulsion film is not yet known. We shall, however, disregarding the molecular constitution of the product, attempt to find a law according to which the density of the film produced varies with the number of α -particles incident upon it.

Table IVA.

Plate, "Instantaneous;" pressure, 747.8 mm.; temperature, 16° C.;
temperature of development, 15°·5 C.; $n_0 = 1.925 \times 10^5$.

No.	Exposure.						Mean intensity, N × 10 ⁻⁸ .	Photometric* reading.	Density, D.
	From—			To—					
	h.	m.	s.	h.	m.	s.			
1	0	8	30	0	28	30	1.872	8.58	3.30
2	0	29	0	0	49	0	1.340	13.0	2.933
3	0	54	30	1	14	30	0.896	4.29	2.406
4	1	14	50	1	29	50	0.462	11.88	1.507
5	1	30	10	1	45	10	0.332	17.17	1.173
6	1	52	50	2	8	0	0.198	25.91	0.779
7	2	8	20	2	18	20	0.0965	36.54	0.412
8	2	18	40	2	28	40	0.0745	39.00	0.336
9	2	29	0	2	38	0	0.0529	42.05	0.222
10	2	38	20	2	46	25	0.0370	43.87	0.187
11	2	46	40	2	52	40	0.0222	46.02	0.121
12	2	53	0	2	57	0	0.0134	47.70	0.070
13	2	57	20	2	59	20	0.00598	Negative spoiled	—
14	2	59	40	3	0	40	0.00296	49.65	0.010
15	3	1	0	3	1	35	0.00164	Too small for measurement	—

* The zero point of the instrument was 50, and the fog density, which is about 0.07, was automatically eliminated. No. 1 and No. 2 were measured by using No. 4 as a supplementary density.

Table IVB.

Plate, "Ordinary;" pressure, 762 mm.; temperature, 16°·5 C.;
Temperature of development, 16° C.; $n_0 = 1.981 \times 10^5$.

No.	$N \times 10^{-8}$.	D.
1	1.932	1.633
2	1.522	1.411
3	0.900	1.160
4	0.610	0.854
5	0.420	0.622
6	0.226	0.379
7	0.110	0.210
8	0.0538	0.121
9	0.0292	0.061
10	0.0186	0.024
11	0.00986	Too small for measurement.
12	0.00661	
13	0.00255	
14	0.00140	

Let m_1 be the amount of silver remaining in its initial state M_1 , in unit area of the film when N α -particles have fallen on it. The silver halide is

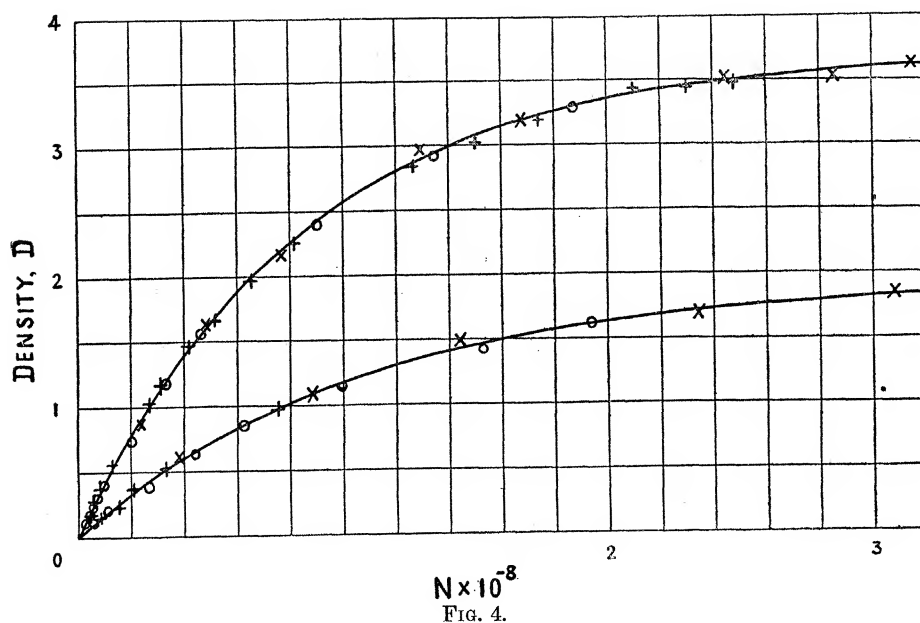


FIG. 4.

imbedded in gelatine in the colloidal state, and we assume that each of these halide grains, when struck by a certain number of α -particles, changes to M_2 , which is different from M_1 , either physically or chemically, and which is more susceptible to development than M_1 . If the halide grains are initially uniformly distributed in the film, the rate of change from M_1 to M_2 , *i.e.* the decrease of m_1 , will be proportional to the amount of M_1 which remains unchanged. Thus

$$-dm_1 = c_1 m_1 dN, \quad \text{so that} \quad m_1 = m_0 e^{-c_1 N},$$

where c_1 is a constant and m_0 is the initial value of m_1 . M_2 might, on further bombardment by α -particles, change to M_3 , differing from M_2 , either chemically or physically, and having a different reducibility. The number of α -particles required to effect the change from M_2 to M_3 would not necessarily be the same as that required to effect the change from M_1 to M_2 .

On the above assumptions, these changes may be represented analytically in an exactly similar way to the successive changes in radio-active substances. If m_2 represents the amount of silver in the state M_2 when N α -particles have fallen on the film, then

$$m_2 = m_0 \frac{c_1}{c_1 - c_2} (e^{-c_2 N} - e^{-c_1 N}), *$$

where c_2 is a constant of M_2 just as c_1 is a constant of M_1 , c_2 probably being

* 'Radio-activity,' 2nd edition, p. 331.

very small compared with c_1 . If the films are simultaneously developed, then

$$D = D_{\max} \frac{c_1}{c_1 - c_2} (e^{-c_2 N} - e^{-c_1 N}).$$

D first increases with N and then, after attaining a maximum value at $N = \log \left(\frac{c_1}{c_2} \right)^{\frac{1}{c_1 - c_2}}$, it decreases.

If $c_2 = 0$, *i.e.* M_2 does not undergo any further change by the α -particles, the expression for D becomes

$$D = D_{\max} (1 - e^{-c_1 N}).$$

This curve is represented graphically in fig. 4, using the following values for the constants :—

$$D_{\max} = 3.7, \quad c_1 = 1.181 \times 10^{-8} \text{ for "Instantaneous" plate,}$$

$$\text{and} \quad D_{\max} = 1.94, \quad c_1 = 0.924 \times 10^{-8} \text{ for "Ordinary" plate.}$$

It will be seen from fig. 4 that the values of the observed densities lie well on the theoretical curves deduced from the above assumptions.

In one experiment the steel rod G was exposed to the radium emanation (the activity of which was equivalent to about 100 milligrammes of radium) and the first photograph was taken exposing the film for three hours to the α -particles emitted. For the next one double the exposure was given and for the third three times the exposure. However, no phenomena of reversal was observed.

In the case of light, if E is the exposure necessary to produce unit density in a film, the exposure required to effect a reversal of density is of the order of $E \times 10^5$, although this depends on the quality and thickness of the film and also on the development. The number of α -particles required to produce unit density in the film "Instantaneous" employed was about 2.7×10^7 per square centimetre, and in the above experiment, although such active emanation was used and the process repeated, the number of α -particles falling on the film was only 1.0×10^9 sq. cm., *i.e.* about 37 times the original value. Consequently this is not a proof that a reversal of density may not be produced by making N sufficiently great. However, it is evident that c_2 is exceedingly small compared with c_1 , if indeed it has any existence, and the experimental results agree with the expression deduced above,

$$D = D_{\max} (1 - e^{-c_1 N}).$$

The maximum density depends on the quality of the film and is proportional to the thickness of the film so long as the α -particles can penetrate it. To produce unit density in the film "Instantaneous" employed, 2.66×10^7

α -particles per square centimetre are required and in the film "Ordinary" 7.8×10^7 per square centimetre. The least number capable of producing an effect which is visible to the naked eye is about 1×10^5 per square centimetre for the former and 1.5×10^5 for the latter.

The Nature of the Photographic Action of α -Particles.

We shall now determine the number of α -particles required to change one silver halide grain. Differentiating the equation

$$m_2 = m_0(1 - e^{-c_1 N}),$$

and omitting the suffixes, we have

$$\delta m = c(m_0 - m) \delta N.$$

Microscopic examination of the film showed that the size of silver grains was independent of the number of the α -particles, but could be increased up to a certain limit (which depends on the quality of the emulsion) on prolonging the time of development. Therefore we may assume for a first approximation that the mass of each grain μ^* on complete development is constant and that the number of the grains s is proportional to m . Thus

$$m = \mu s \quad \text{and} \quad m_0 = \mu s_0,$$

where s_0 is the number of the grains which would be present in unit area of a film of the maximum density. Substituting these values in the above equation, we have

$$\delta s = c(s_0 - s) \delta N.$$

If we take the corresponding values of s and N in a film of very small density, s is negligibly small compared with s_0 , and therefore $\delta s = cs_0 \delta N$.

In order to determine the number of α -particles required to change one halide grain, the value of cs_0 in the above equation must be obtained. It has already been shown that the values of c from Curves I and II, fig. 4, are

$$c = 1.181 \times 10^{-8} \text{ for the plate "Instantaneous,"}$$

$$c = 0.924 \times 10^{-8} \text{ for the plate "Ordinary."}$$

The direct counting of individual grains over a small area 1.225×10^{-5} sq. cm. of plates was done by means of a microscope of 700 diameters. It was found impossible to determine s_0 directly, as the grains were too densely packed in the film for counting, but by taking a plate upon which a small number of α -particles had been allowed to fall, the counting presented no great difficulty. By making use of the relation $s_0 = s \frac{m_0}{m} = s \frac{D_{\max}}{D}$, s_0 was at once determined.

* Actually μ may be regarded as the average mass of the grains.

The photograph produced by the α -particles was a dark circular area, outside which was a portion of very slight density due to γ -rays and probably to fog. These two factors must also have affected the darker area, and allowance has been made for them, as may be seen in the following examples.

Plate "Instantaneous," No. 7, given in Table IVa.

$$D = 0.412.$$

The number of grains in 1.225×10^{-5} sq. cm. of dark area	210
" " " " outside dark area ...	52
Difference (due to the α -particles)	158

$$s_0 = \frac{158}{1.225 \times 10^{-5}} \times \frac{3.7}{0.412} = 1.16 \times 10^8 \text{ per sq. cm.}$$

Plate "Ordinary," No. 7; given in Table IVb.

$$D = 0.210.$$

The number of grains in 1.225×10^{-5} sq. cm. of dark area	148.5
" " " " outside dark area ...	20.0
Difference (due to the α -particles)	128.5

$$s_0 = \frac{128.5}{1.225 \times 10^{-5}} \times \frac{1.94}{0.21} = 0.97 \times 10^8.$$

Therefore $cs_0 = 1.37$ for the plate "Instantaneous,"
and $cs_0 = 0.90$ for the plate "Ordinary."

Thus cs_0 is a quantity of the order unity for both films, and the increase in the number of grains is nearly equal to that in the number of the α -particles. This at once points to the conclusion that a halide grain may be changed when struck by a single α -particle.

To ascertain the actual number of α -particles which struck each halide grain, we must know the distribution of the halide grains in the emulsion film and also their dimensions, since:—

(1) Some α -particles might pass through the film without striking any of the halide grains;

(2) Some α -particles might strike more than one halide grain;

(3) Or some halide grains might be struck by more than one α -particle.

The following considerations, however, throw some light on these points:—

(1) The "Instantaneous" film of very high density is entirely covered with silver grains. Therefore, it was not likely that the α -particles passed through the film without striking any halide grains.

The "Ordinary" film of maximum density was *almost* covered with silver

grains, about 10 per cent. of the area being estimated as allowing α -particles to pass through without contact with the grains.

(2) In the "Instantaneous" film of small density about one-third of the grains were overlapping others. Therefore, on the average about 30 to 35 per cent. of the α -particles each struck two halide grains in passing through the film. In the "Ordinary" film the overlapping was negligible, so that one α -particle did not strike more than one halide grain.

(3) In these films of very small density in question the grains are very diffusely distributed and it is improbable that one halide grain would be struck by more than one α -particle.

From the above facts, it can be seen that in the "Instantaneous" film an α -particle always struck at least one halide grain, and one-third of the α -particles on the average struck two halide grains. The "Ordinary" film nearly fulfilled the condition that one α -particle should always strike one and only one halide grain. The deviation of cs_0 from unity being thus explained by the above consideration, it is clear that a halide grain becomes developable when struck by one α -particle.

The numbers of silver grains per square centimetre (or, in cases of overlapping, groups of grains) counted by means of the microscope are given below for comparison with the number of α -particles incident upon the same area.

An "Ordinary" plate (a part of the above-mentioned plate) exposed to light and similarly developed was examined under the microscope. The number of grains per unit area in this film was found to be greater than that in the film of the same density which had been exposed to α -rays, and the variation in size of the grains was much greater. This shows that the above results are not accidental.

Table VA.—Plates "Instantaneous" given in Table IVA.

I	II	III	IV	V	VI
No.	Number of grains in 1.225×10^{-5} sq. cm. of dark area.	Number of grains in 1.225×10^{-5} sq. cm. outside dark area.	Difference.	Number of grains per sq. cm. (due to α -particles).	Number of α -particles incident upon the film per sq. cm.
7	173	52	121	9.9×10^6	9.65×10^6
8	142	52	90	7.35×10^6	7.45×10^6
9	107	47	60	4.9×10^6	5.29×10^6
10	75	32.5	42.5	3.47×10^6	3.12×10^6
11	71	42.4	28.6	2.33×10^6	2.02×10^6
12	67	52	15	1.22×10^6	1.22×10^6

Table VB.—Plates “Ordinary” given in Table IVB.

I	II	III	IV	V	VI
No.	Number of grains in 1.225×10^{-8} sq. cm. of dark area.	Number of grains in 1.225×10^{-8} sq. cm. outside dark area.	Difference.	Number of grains per sq. cm. (due to α -particles).	Number of α -particles incident upon the film per sq. cm.
7	148.5	20	128.5	10.5×10^6	11.0×10^6
8	89.8	23.6	66.2	5.4×10^6	5.37×10^6
9	55.1	20.5	34.6	2.82×10^6	2.92×10^6
10	37.3	14.3	23.0	1.88×10^6	1.85×10^6
11	25.7	14.0	11.7	0.955×10^6	0.986×10^6
12	23.1	14.6	8.5	0.694×10^6	0.661×10^6

We are now in a position to elucidate the actual mechanism of the photographic process effected by α -particles.

If α -particles travel through a gas, they ionise the gas, *i.e.* break up the gas molecules into ions. The ions thus formed recombine amongst themselves or, if an electric force be applied, produce a current. From the experiments of Bragg and Kleeman,* and recently of Geiger, it is known that the ionisation in gases per unit path increases at first, reaches a maximum (about 3.5 times the initial value) near the end of the range (about 93 per cent. of the total), and then rapidly diminishes to zero.† Since the absorption in a gas is due to ionisation, it is reasonable to suppose that the absorption in a solid, which is of the same character as in a gas, is due to ionisation of a similar type. The reason that the ionisation of the silver halide, under this circumstance, is not followed by recombination is probably due to the fact that the surrounding medium, *e.g.* the gelatine, absorbs the liberated halogen ions.‡ We shall now show that the ionisation of the above-mentioned type in the emulsion film can produce the photographic action, which, as we have observed, remains *constant* as the distance traversed by the α -particles increases over a certain range (about 96 per cent. of the total) and then rapidly diminishes to zero.

It has been shown that the number of grains depends only on the exposure, *i.e.* on the number of α -particles falling on the film. Moreover, the size of the grains increases to a certain limit on prolonged development, which depends on the quality of the film but *not* on the number of the

* Bragg and Kleeman, *loc. cit.*

† Geiger, ‘Roy. Soc. Proc.’ May, 1909.

‡ Abney, ‘Photo. News,’ Aug., 1882; H. B. Baker, ‘Chem. Soc. Journ.’ p. 728, 1892; Hitchcock, ‘Amer. Chem. Journ.’, vol. 11, p. 474; vol. 13, p. 273.

α -particles. These experimental facts show that, if some of the halide molecules within a grain are initially ionised by an α -particle, the whole grain becomes capable of development, but the reduction cannot extend to other grains which have not been initially ionised. Suppose an α -particle penetrates n halide grains centrally, one after the other, then the magnitude of ionisation in each grain is not the same, but increases one after the other and is a maximum in the grain occupying a position nearly at the end of the path of the α -particle. When, however, the film is completely developed, all the grains are reduced to a constant limit, which is *independent* of the primary ionisation, and depends on the size of halide grains in the emulsion film, and thus we obtain silver grains as a secondary consequence of the ionisation, so that the photographic action is constant throughout the range of the α -particle in the substance. This explains why the photographic action fails to exhibit the characteristic features of the ionisation curve to which reference has just been made. The reason why the photographic action is not constant up to the end of the total range can be explained from the fact, of which I was informed by Dr. Geiger, that a fraction of the α -particles is already stopped a few millimetres before the actual end of the range.

Let us now interpret the constant c , which enters into the expression $D = D_{\max}(1 - e^{-cN})$ and characterises the photographic films.

We have deduced the following equation from the above—

$$\delta s = c(s_0 - s) \delta N,$$

and proved that each halide grain is rendered capable of development when struck by an α -particle. Consequently c is the probability of one halide grain* being struck by an α -particle, and therefore will be the ratio of the area of the projection of each halide grain to a plane perpendicular to the direction of the motion of the α -particle to 1 sq. cm.

If r be the average radius of the halide grains, supposing these to be spherical, then

$$c = \pi r^2 / 1.$$

Consequently

$$r = \sqrt{\frac{c}{\pi}} = \sqrt{\frac{1.181 \times 10^{-8}}{\pi}} = 6.13 \times 10^{-5} \text{ cm. for the plate "Instantaneous,"}$$

and

$$r = \sqrt{\frac{c}{\pi}} = \sqrt{\frac{0.924 \times 10^{-8}}{\pi}} = 5.42 \times 10^{-5} \text{ cm. for the plate "Ordinary."}$$

To test this assumption we may deduce the mass of silver per square

* Strictly speaking, the sphere of action must be taken, but the grains being exceedingly large compared with molecular dimensions, the error due to this cause will not affect the calculation to estimate the approximate order of magnitude.

centimetre of a film containing a known number of grains and compare it with that calculated from its photometric density (amount of silver per square centimetre in gramme* = $1.03 \times 10^{-4} \times D$).

Table VI.

	Plate No. 7, "Instantaneous," given in Table IVA.	Plate No. 7, "Ordinary," given in Table IVB.
r , radius of halide grains = $\sqrt{\frac{c}{\pi}}$	6.13×10^{-5}	5.42×10^{-5}
V , volume occupied by halide grains = $\frac{4\pi}{3} r^3 \times \text{number}$ of grains per sq. cm.	1.25×10^{-5}	0.702×10^{-5}
Mass of silver bromide per sq. cm. = $V \times 6.33$ in gramme	7.88×10^{-5}	4.44×10^{-5}
Mass of silver per sq. cm. = $V \times 6.33 \times \frac{108}{188}$ in gramme	4.53×10^{-5}	2.55×10^{-5}
Mass of silver calculated from density = $D \times 1.03 \times 10^{-4}$	4.25×10^{-5}	2.16×10^{-5}

Bearing in mind the fact that the conversion of silver halide to metallic silver is not carried to completion, the agreement between the values obtained by the two methods is seen to be quite satisfactory.

Photographic Action of β- and γ-Rays.

In the experiments described, we have β - and γ -rays accompanying the α -rays, and some experiments were accordingly performed to determine the photographic effect of the former. When the rays (for apparatus see figs. 1 and 3) were screened by a sheet of aluminium, the thickness of which was just sufficient to cut off the α -rays, no appreciable density was observed in the film for exposures which, in the case of α -rays, would produce densities of 2 or 2.5. On using a very active deposit of radium C, and giving a sufficiently long exposure, the exposed part of the plate became darkened, but there was no sharp line of demarcation, showing the great penetrating power of the γ -rays. Thus, the action of γ -rays on the film employed is seen to be negligibly small compared with that of the α -rays.

It must be remembered that we are not speaking of the total power of γ -rays in affecting the film. On account of their great penetrating power, the range of the photographic action of the γ -rays is several thousand times greater than that of the α -rays.

The photographic action of β -rays in the film employed was about one-third or one-quarter of that of α -rays.

* Sheppard and Mees, *loc. cit.*

Since β -rays are more penetrating than α -rays, the total power of affecting a photographic film may be greater than that of the α -rays.

It is difficult to compare quantitatively the photographic action of β - and α -rays, the initial velocities and laws of absorption being different.

The Variation of the Film Density with—

- (1) The number of incident α -particles.
- (2) The intensity of the incident light.

While the density produced in a film on development can be expressed in terms of the total number of α -particles incident upon the film, independent of the intensity and the velocity of the rays, the density in the case of light is a function of It^p , as already described. Moreover, the density in the latter case depends on the wave-length of the light. Hurter and Driffeld* obtained a relation between the density and the exposure on the assumption that the rate of the change of silver halide in a film, while being exposed to light of intensity I , is proportional to the amount of light absorbed by the halide not yet affected, as follows :

$$D = \gamma \log \{e^{km_0} - (e^{km_0} - 1)e^{-\frac{It}{iek m_0}}\},$$

where γ is the development factor, *i.e.* a constant, m_0 the total amount of the halide per unit area initially present, and k the coefficient of the absorption of the halide, being determined by its amount and having the same value for the changed and the unchanged substance. Disregarding the reversal of the density, this equation is in good agreement with the experiments for a considerable range of exposure, extending to so-called under- and over-exposures (exposures of from about 1 to 10,000 C.M.S.).

An approximate form of this equation is

$$D = \gamma \log \left(\frac{It}{i} \right),$$

from which we speak of the "Inertia" i of a plate.

The expression of the density being thus fundamentally different, these terms cannot characterise a photographic plate for α -particles. Suppose we have two emulsion films I and O of equal weight and similar composition, the only difference between them being that the size of halide grains in I is greater than that in O. Then the inertia of film I would be less than that of O, and consequently the former is more sensitive to light than the latter. Suppose these two films are equally exposed to α -particles and developed under identical conditions. The numbers of halide grains per unit area in two films are as

$$r_I^{-3} \quad \text{to} \quad r_O^{-3},$$

* 'Journ. Soc. Chem. Ind.,' May, 1890.

where r_I and r_O are the average radii of the grains in films I and O respectively, assuming they are spherical. If the grains are uniformly distributed in the films, then the probabilities of any one halide being struck by an α -particle per unit area, being proportional to the sum of the areas of the diametrical sections of the halide grains per unit area perpendicular to the direction of the motion of the α -particle, are as

$$r_I^2 \cdot r_I^{-3} \text{ to } r_I^2 \cdot r_O^{-3}, \text{ or as } r_I^{-1} \text{ to } r_O^{-1}.$$

Therefore, on the average, the number of grains struck by a certain number of α -particles is greater in O than in I. But, since the mass of each grain is proportional to r^3 , the total mass of the grains changed are as

$$r_I^2 \text{ to } r_O^2.$$

Thus, if we speak of the density, I is more sensitive than O, but if we speak of the number of grains, O is, on the contrary, more sensitive than I.

It will be noticed from Tables VA and VB that the number of grains due to γ -rays, and also probably due to the fog in the film "Ordinary," is much less than that in the film "Instantaneous" for the same number of α -particles. It is therefore hoped, by preparing an emulsion film* of very fine halide grains, and by using a microscope of high magnification, that the photographic method can be applied for counting α -particles with considerable accuracy.

In conclusion, I wish to express my sincere thanks to Prof. Rutherford for suggesting the investigation, and for his continual interest during its progress at the University of Manchester. I must also thank Mr. C. W. Gamble, M.Sc.Tech., of the Municipal School of Technology, for his great kindness in allowing me to use the König's spectrophotometer, and for much valuable advice in carrying out the photographic operations.

* The film must be so thick that no α -particles are allowed to pass through the film without contact with the grains.